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The invention relates to adhesiveless groups from Polyarylenetherketonen, Polyphenylensulfiden or thermoplastic polyesters if necessary with further group components, as well as a method to the preparation of these groups.

Laminates of polyester foils and mica paper and/or Ceramic(s) fiber papers, which stuck together by adhesives, for example z become as isolating foils in the electrical industry. B. for engines, generators and transformers uses. A substantial disadvantage of these laminates is the presence of an adhesive layer, which must be laid on in a separate processing step and which represents a weak point in the laminate.

Task of the invention was it to manufacture groups with improved properties after a simple method. Those gave up by the fact is solved that shaped parts from Polyarylenetherketonen, Polyphenylensulfiden or thermoplastic polyesters are injected at temperatures between their glass transition temperature and their melting point with the other group components.

Subject-matter of the invention are therefore groups, if necessary which consist of shaped parts of Polyarylenetherketonen (PAEK) and/or Polyphenylensulfiden (PPS) and/or thermoplastic polyesters (PES) and further shaped parts, which are available by injecting the shaped parts at temperatures between the glass transition temperature (Tg) and the melting point of the PAEK, PPS or PES. The groups can consist either of shaped parts of same materials or of different materials, which are connected in two or several situations.

top A substantial advantage of the groups according to invention is in the fact that they consist only of that to connecting components and no kind-strange materials for sticking the individual coatings together are necessary. The manufacturing of the groups according to invention succeeds already at temperatures under the melting point of the PAEK, to PPS and/or. PES. This brings the advantage in relation to the well-known fusion sticking procedures that the shaped

parts which can be connected remain not to have not to be melted and thus both its inherent stability and even thickness, and its electrical and mechanical properties. In addition an additional advantage is in it that due to the substantially lower temperatures when injecting the preparation is substantially smaller more simply and the energy consumption.

The groups are preferred at temperatures between Tg and (Tg + 100 DEG C), particularly preferentially between (Tg + 10 DEG C) and (Tg + 60 DEG C) injects. With the help of the method according to invention it is also possible, the tackness and/or. Adhesion of the PAEK to steer PPS and PES shaped parts and thus the adhesion of the group parts according to the requirements to the groups. Like that is the adhesion and/or. Adhesion between the parts for example when injecting with the Tg less solid, it, which can be connected, rises up to a temperature of approximately (Tg + 10 DEG C) to approximately (Tg + 100 DEG C) to a maximum, in order to then drop up to the melting point again. This is of advantage above all, where a defined adhesion is necessary. For example is with certain electrical laminates, as so-called safety isolation of several single situations it consists which necessarily for inspection purposes a non-destructive Delaminierbarkeit and thus a relatively small adhesion. On the other hand for example a particularly good adhesion is necessary for standard electrical laminates.

A further possibility for adjustment a defined adhesion consists of varying the press time or the pressing power whereby a short press time or a small pressing power causes an accordingly smaller adhesion of the group parts. The pressing power is usually with approximately 0.5 to 4 bar. Particularly effectively the tackness and adhesion in the group D can become the relationship of the crystalline and amorphous portion in the used PAEK, PPS and PES shaped parts controlled. Thus becomes for example with use of PAEK, PPS or PES films with small crystalline portion (for example 0 to approx. 15% crystalline portion), which will receive by rapid cooling of the melt during the foil production, a substantially stronger tackness and adhesion of the groups reach, than with use of films with higher crystalline portion (for example approx. 15 to approx. 30% crystalline portion), which will receive by slow cooling of the melt during the foil production or by subsequent annealing of the film. For the achievement of connecting with good adhesion prefers high-amorphous shaped parts from PAEK, PPS and/or PES with a crystalline portion from approximately 0 to 15% used.

As PAEK, PPS and PES shaped parts are in particular films, tapes or plates, however also three-dimensional parts such as z. B. Deep-drawing article, injection moulding article, to understand tubes or cases. As further shaped parts, which are along-injected if necessary with the preparation of the groups, come in particular inorganic or organic papers, for example mica papers, era avoiding papers (z. B. Nomex TM 411 of you Pont), papers on base of cellulose, aluminium oxide or magnesium oxide (z. B. Tufquin TM, and/or. Cequin TM, of companies Quin t, the USA; inorganic or organic fleeces, fabrics or thread clutches of eggs, for example on base of natural or synthetic textile fibers, glass fibers or carbon fibers; Metal fabric, metal foils, z. B. Kupferfolien; Plastic foils; as well as inorganic o of organic pigments and powders, like z. B. Kieselsäure (SiO₂) or calcium carbonate (CaCO₃) in question. PAEK are commercially available, for example Polyetherketone (PEK) as Ultrapek TM (BASF), Polyetheretherketone

(PEEK) as Victrex TM (ICI), Polyetherketonketone (PEKK) of you Pont, Polyetheretherketonketone (PEEKK) as Hostatec TM (most). PPS is likewise commercially available, for example as Fortron TM (most) or Ryton TM (Phillips). As PES for example polyethylene terephthalate (PETP), z come. B. Melinex TM von Fa. ICI or polybutylene terephthalate (PBTP), z. B. Vestodur TM von Fa. Hüls in question. PAEK and/or. PPS are for example in the DE-PS 39 31 649 or US 49 96 287 described.

Table 1

Glass transition temperatures (Tg) and melting points (TM) in DEG C
EMI4.1

The processing of the PAEK, PPS and/or. PES to shaped parts effected after well-known methods, for example via extruding, deep-drawing, powder pressing or injection moulding. The preparation of films succeeds for example through to melt opens of the granular materials in the extruder, extruding by a nozzle and a cooling of the melt in the water bath or on a casting roll. Can be increased by rapid cooling the amorphous portion polymers. Slow cooling by higher continuous casting and rolling temperature and/or. subsequent annealing lead to an increase of the crystalline portion in the film. In this way it is possible, by appropriate choice of the cooling conditions and/or. to regulate by following annealing the crystalline portion in the shaped parts. Films from PAEK, PPS and PES are also commercially available, for example PEEK films or PPS films as Litrex TM with PCD Polymere. Die making of PAEK films are for example in US 49 96 287 described.

The making of the groups according to invention can take place both on stationary pressing, and, in particular for bahnförmige groups, continuous, for example on Laminatoren, calenders or double volume presses. For continuous laminating the preheating of the courses is recommended and/or. Films on heating rollers, in a heat tunnel or with IR beams before the intake into the laminating plant. In case of the use to films their thickness is usually with approximately 0.005 to 1 mm, preferentially about 0,025 to 0.2 mm. The advantage of continuous laminating is above all in the fact that also multilevel laminates with most different substrates can be manufactured in simple and energy-saving way in a run. With the help of the method according to invention it is further also possible to manufacture tubes. This takes place for example with spiral wrapped tubes via the fact that films spiraled are overlapping wound and injected. The making of pigment or powder groups takes place for example via applying of the pigments or powders on PAEK, PPS or PES films or - shaped parts, for example by shaking filters or spray guns, and afterwards injecting. The pressing power depends on the desired adhesion of the group parts and is usually about 0,5 to 4 bar. Further it can be of advantage to roughen up or several of the shaped parts before injecting at the surface, which can be connected for example by mechanical roughening, chemical up etching, plasma corrodung or Coronavorbehandlu

In the examples the following shaped parts were injected with one another: PEEK film:

Litrex TM K, thickness: 0.05 mm

crystalline portion: 7% (PCD of polymers) PPS film:

Made of PPS (Ryton TM, Phillips) by extruding with 330 DEG C over a broad slot nozzle on a casting roll with 20 DEG C

Thickness: 0.05 mm, crystalline portion: approx. 7% polyester foil:

Made of PETP (polyethylene terephthalate, Melinex TM, ICI) by extruding with 300 DEG C on a casting roll with 10 DEG C

Thickness: 0.05 mm, crystalline portion: approx. 10% era avoiding paper:

Nomex TM 411, 41 g/m² (You Pont) era avoiding paper with mica:

Nomex TM 418, 150 g/m² (You Pont) mica paper:

75 and/or. 180 g/m² (Companies Samica, UDD) of isolating papers:

Tufquin TM 110 and 120, 90 and/or. 130 g/m² (Companies Quin t, the USA)

Cequin I and 3000, 140 g/m² (Companies Quin t, the USA) mat:

50 g/m² (Companies Schuller, FRG) glass fabric:

30 g/m² (Fa.Schuller, FRG) Kupferfolie:

0.017 mm (companies Gould, England)

The identification of the crystalline portion (crystallinity degree) took place by means of DSC (differential scanning calorimetry).

The adhesion between the individual shaped parts in the groups was determined by the peeling firmness in accordance with DIN 53282. When measure for the adhesion serves the force (N/15 mm), which is necessary for the Delaminierung.

Example 1

A press with a press surface of 300,300 mm was preheated on 160 DEG C. A mica paper and a PEEK film were laid on top of each other, inserted between two aluminum plates made antiadhesively with parting agent into the press and injected during 10 seconds with a pressure by 2 bar. Subsequently, the group was inferred from the press and cooled down on ambient temperature. The group showed good adhesion. With the attempt, the group to delaminieren the adhesion between mica paper and PEEK film was larger than the co-operation of the mica paper.

Examples 2 to 15

Analog for example 1 groups, whereby however the films stated in table 2, fleeces were manufactured and/or. Papers were used. Well in each case responsible groups were received. The peeling firmness of the Aramidpapier/PEEK group in accordance with example 4 is with 2 N/15 mm. The peeling firmness of the PEEK/PEEK group in accordance with example 15 is with 25 N/15 mm.

Examples 16 to 18

Analog for example 1 groups were manufactured, whereby however, as in table 2, PPS films with a PEEK film, with mica paper and with era avoiding paper were stated were injected. The press temperature was with 120 DEG C. Well in each case responsible groups were received. The peeling firmness of the PEEK/PPS Verbun in accordance with example 16 was with 27 N/15 mm. Table 2: Groups example 1 Glimmerpapier/PEEK film

- 2 Glimmerpapier/PEEK Folie/Aramidpapier
- 3 Glimmerpapier/PEEK Folie/Glimmerpapier
- 4 Aramidpapier/PEEK Folie/Aramidpapier
- 5 era avoiding paper with Glimmer/PEEK film
- 6 era avoiding paper with Glimmer/PEEK Folie/Aramidpapier with mica
- 7 Tufquin TM 110/PEEK-Folie
- 8 Tufquin TM 120/PEEK-Folie
- 9 Cequin TM I/PEEK film
- 10 Cequin TM 3000/PEEK-Folie
- 11 Glasvlies/PEEK film
- 12 Glasgewebe/PEEK Folie/Glasgewebe
- 13 Kupferfolie/PEEK film
- 14 Kupfergewebe/PEEK film
- 15 PEEK Folie/PEEK film
- 16 PEEK Folie/PPS film
- 17 Glimmerpapier/PPS film
- 18 Aramidpapier/PPS film
- 19 Glimmerpapier/PETP Folie/Glimmerpapier
- 20 Aramidpapier/PETP film
- 21 PETP Folie/PETP film

Examples 19 to 21

Analog for example 1 groups were manufactured, whereby however as in table 2, PETP films with mica paper, with era avoiding paper and with second PETP films were stated were injected. The press temperature was with 100 DEG C. Well in each case responsible groups were received. At the group in accordance with example 20 the adhesion between era avoiding paper and PETP film was larger than the co-operation within the era avoiding paper. The era avoiding paper tore up already with 0,8 N/15 mm.

Example 22

Analog for example 4 a group was made of Aramidpapier/PEEK film/era avoiding paper, however with the difference that the press temperature was with 146 DEG C. The group showed a smaller adhesion than those in accordance with example 4 and could easily be delaminieren. The peeling firmness is with 0,1 N/15 mm.

Example 23

With the help of a shaking filter calcium carbonate powder became (MILLICARB 0.003 mm, companies Plüss Stauffer, Switzerland) in a quantity of 3 g/m² on a PEEK film applied and afterwards analog for example 1 with 160 DEG injects. A g responsible group was received.

Example 24

In order to point the influence out of the press temperature on the peeling firmness of the groups, analog for example 1, however at the temperatures, was manufactured as well as with a pressure by 2 bar and a press time by 2 seconds, indicated in table 3, a PEEK/PEEK group as well as a PEEK/Aramidpapier group. The values of the peeling strengthenesses in N/15 mm are likewise in table 3 arranged.

Table 3

Peeling firmness of the groups (N/15 mm)
EMI11.1

The low peeling strengthenesses of the groups with era avoiding paper arises as a result of the fact that the era avoiding paper tears up with approximately 0.7 N/15 mm, although it sticks still to the film. The adhesion at the film is thus larger than the internal co-operation of the era avoiding paper. The relatively high value of 1,8 N/15 mm with 300 DEG C comes off above all by the fact that the era avoiding paper is partly pressed into already the PEEK film relatively soft at this temperature, whereby it comes to an additional mechanical anchoring.

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1. Groups consisting of shaped parts of Polyarylenetherketonen (PAEK) and/or Polyphenylensulfiden (PPS) and/or thermoplastic polyesters (PES) and if necessary further shaped parts, which are available by injecting the shaped parts at temperatures between the glass transition temperature (Tg) and the melting point of the PAEK, PPS or PES.
2. Groups according to claim 1, characterised in that the shaped parts at temperatures between Tg and (Tg + 100 DEG C) to be injected.
3. Groups according to claim 2, characterised in that the shaped parts at temperatures between (Tg + 10 DEG C) and (Tg + 60 DEG C) to be injected.
4. Groups in accordance with one of the claims 1 to 3, characterised in that the shaped parts from PAEK, PPS and/or PES a crystalline portion from 0 to 15 96 exhibit.
5. Groups in accordance with one of the claims 1 to 4, characterised in that the further shaped parts from inorganic or organic papers, fleeces, fabrics or thread clutches of eggs, metal foils, plastic foils, inorganic or organic pigments or powders exist.
6. Methods to the making by groups in accordance with one of the claims 1 to 5, that shaped parts from PAEK and/or PPS and/or PES and further shaped parts, characterized thus, at temperatures between Tg and the melting point of the PAEK, PPS or PES with one another are if necessary injected.

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